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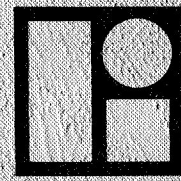
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INVESTIGATION OF SPUTTERING EFFECTS ON
THE MOON'S SURFACE

Fourteenth Quarterly Status Report
Contract NASw-751

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ABSTRACT

The possibility that darkening of powder samples in laboratory ex-
periments with a simulated solar wind is due to carbon contamination is
re-examined and shown to be probable. A suggestion by D. Nash (to be
published) that surface catalysis affects the amount of darkening arising
from carbon contaminants is tentatively accepted. Catalysis is cited as
the probable cause for greater darkening of certain powder mixtures than
of the separate pure powders. Darkening effects due to carbon at the
lunar surface are evaluated semi-quantitatively. The input of carbon
from the solar wind alone is shown to be capable of darkening up to
roughly 400 gm/cm^2 of a lunar surface powder. The observation in Sur-
veyor 1 photographs of high albedo rocks amidst a dark powder that grows
darker with depth is presented as evidence that darkening actually occurs
for material exposed at the lunar surface. An alternative hypothesis
that exposed surfaces increase in albedo (bleaching) is examined and re-
jected. A model of surface powder formation including darkening is pre-
sented and generalized to other lunar features.

The fact that the dependence of the brightness of a number of lunar
features upon the angles of incidence and emergence is governed by a
Lommel-Seeliger factor is reinterpreted. The dependence is only approxi-
mate and must fail near the illuminated limb and at large phase angles.

The slight change in the apparent lunar color at phase angles up to 60°
is suggested to result from multiple scattering. Multiple scattering
is suggested as the cause of an abrupt change in lunar or powder sample
brightness at phase angles smaller than 3° ("opposition effect").

I. INTRODUCTION

In this quarter we received a very interesting paper by D. Nash titled "Proton-irradiation darkening of rock powders and solar-wind darkening of the Moon", which he submitted to the Journal of Geophysical Research. Later we received another paper submitted to the same journal by R. Greer and B. Hapke titled "Electron microprobe-characterization of microcrystalline powders darkened by simulated solar-wind irradiation". Since we had also submitted a paper to that journal on a closely related subject, namely, the optical properties of powders darkened by a simulated solar wind, the editor suggested publication of the three papers together, with opportunity for comments on the parallel papers. Our revised paper has now been submitted to the editor.

The revisions, together with supporting measurements and calculations, constituted a major effort of this quarter and form the latter portion of this report. The full paper will be included in a later report in reprint form. The main revisions involved the powder-darkening controversy, the relation of approximate Lommel-Seeliger scattering by central lunar features to surface roughness and compaction in those features, the "opposition effect" in the brightness of natural surfaces at very small phase angles, and the relation of multiple scattering to slight changes in color of natural surfaces at moderate phase angles. Our original conclusions were not modified.

Studies have continued on the question of water formation due to the solar-wind bombardment of silicates. Stecher and Williams[1] have

published a paper closely related to this topic, "Interstellar molecule formation". They point out that molecule formation under low pressure conditions takes place principally on small solid grains by chemical-exchange reactions. The result in our experiments is that H_2O release follows a gradual buildup of OH concentration at or near the solid silicate surface when bombarded by hydrogen ions. The H_2O emission strongly resembles the emission expected from H_2O desorption that would result from a temperature wave propagating through the powder surface which in turn would result from sample heating by ion bombardment. Efforts are continuing towards a better understanding of these effects.

In conjunction with the water-formation studies, darkening of the samples under better-known vacuum conditions is being studied. Experiments are planned which should test the importance of catalytic cracking of carbon-carrying molecules on the sample surfaces. Other experiments will be devoted to isolating the cause of darkening. For example, we can vary the partial pressures of carbon monoxide, carbon dioxide, water, or pump oil vapor at will. We will attempt to correlate darkening with amounts of these molecules holding the sample conditions fixed.

Another problem that remains is that we have so far failed to find a powder which simultaneously matches the photometric properties of the moon in backscatter and approximate Lommel-Seeliger scattering. A mixture of basalt and greenstone should satisfy the former property because

greenstone backscatters too strongly while basalt is slightly too difuse as a scatterer. We expect that the simultaneous matching of these two properties should have implications for the compaction of the lunar surface powder.

In polarization we want to look at the effects of changes in the index of refraction. Possibly our inference concerning particle sizes in the lunar surface powder would be changed if this variable were introduced.

Beam experiments seem pointless if carbon-carrying background gases must be kept below 10^{-10} Torr (see Section III) as our present beam facility is then inadequate. Decisions on this point will follow from the above experiments.

II. STRATIGRAPHY OF THE LUNAR SURFACE POWDER AND SURFACE DARKENING

Photographs obtained by Surveyor 1 revealed that surface rocks are higher in albedo than a finely divided powder that covers most of the surface. In addition, material from a few centimeters below the local surface was exposed by the motion of the Surveyor footpad upon landing. This disturbed material was darker than nearby undisturbed surface material. We want to point out that these three types of surfaces visible in the Surveyor photographs represent features of different age, in general. We will outline a model of the erosion history in the vicinity of Surveyor 1. The basic suggestion is that a lunar surface soil is being derived from surface rocks in analogy to the terrestrial case. In the case of the moon, however, the erosion mechanism is the bombardment by micrometeorites, rather than weathering. Since both micrometeorites and larger meteoroids stir the rock fragments on the lunar surface, the analysis is complicated. But a semi-quantitative calculation establishes that the rock surfaces are the youngest viewed and the subsurface material is the oldest. The succession of albedos then strongly points to a progressive darkening of exposed surfaces. The model is then generalized to explain some albedo contrasts on the moon.

To clarify the role of lunar surface darkening in this context, let us first suppose that there were no mechanism by which rock fragments on the moon are darkened. When rock material is fragmented, the albedo is invariably increased because less light is lost by absorption within

the volume of the rock particles; more of the light is reflected at particle surfaces. Then the presence of high albedo rocks in the midst of a dark soil becomes an enigma unless we postulate the existence of a mechanism which brightens the surface of the soil slightly and which brightens the rocks substantially. The choice between a darkening and brightening mechanism is not immediately obvious but we will argue that brightening is unlikely and show why the observed darkening is to be expected.

The smallest meteoroids from the interplanetary medium disturb only a superficial layer on the moon, but their number is so great that a unit area of the moon will be completely covered by primary impact craters in some (finite) characteristic time. On the other hand, meteoroid impacts involving masses above some critical size are so rare that the large craters that result remain for times of the order of the age of the moon. Thus the remains of large events on the moon are generally ancient while those from small events are relatively young, as has often been recognized. Put another way, the moon is occasionally stirred to great depth and commonly stirred to shallow depth. The rare events expose deep-lying rock and the interim events tend to reduce those rocks to finer rubble. The rate of surface feature obliteration was considered, for example, by Bastin[2] who concluded that, as a lower limit, primary craters alone of a few centimeters diameter must have completely disturbed a unit area of the moon in the past 10^7 yr. A more realistic influx of meteoric material than the lower limit plus consideration of secondary cratering effects

-7-

could reduce this estimated time scale several orders of magnitude. If this is the case, the possibility exists that ancient rocks of up to roughly 1 m diameter in the Surveyor vicinity may have been reduced completely to a finely divided debris that now tends to bury and protect the remaining rocks.

Most of the surface area around Surveyor 1 consists of a material too finely divided to be resolved (<0.5 mm). Gault et al.[3] have emphasized that the crater shapes in the photographs of Luna 9 and Surveyor 1 require noncohesive or weakly cohesive surface materials having particles not exceeding a millimeter in size. The imprint of the footpad of Surveyor 1 is so smooth that it has been widely recognized that the surface material in that locality has particles mostly much smaller than 1 mm. Only craters deeper than 1 m near Surveyor 1 are clearly associated with the angular blocks that appear to be rocks. Therefore 1 m may well be the approximate depth of the finely divided rock material (soil) near Surveyor 1. The few rocks that are seen in that region seem to be in various stages of burial and erosion. Sharply angular rocks tend to be well-exposed while rounded (eroded?) rocks give the impression of being in the last stages of burial. Rocks of 1 m diameter occupy only 1% of the surface and all fragments at the surface that are visibly angular occupy only 7.6% of the surface.[4] If we consider, then, that 1 m rocks constitute what is now left of the source of surface soil, we see that the present soil source is very dilute.

The material disturbed by the motion of the Surveyor 1 footpad had a visual albedo of 5.7% while the undisturbed surface had an albedo of 7.7%.[4] The rock albedos have not been reported. An albedo of 25% for rock powder is representative in our experience and only a 10% admixture of a powder of 25% albedo in a powder of 5.7% albedo is needed to raise the albedo to 7.7%.

Let us now consider a model of the soil deposition near Surveyor 1. We do not know how deep micrometeorite ejecta from the rocks will penetrate into the surface material nor how deep the micrometeorites will mix this ejecta into the dark material. Either depth is probably of the order of ten times the largest micrometeorite size that we have to take into consideration. Meteoroids up to 100μ diameter, for example, should affect a depth considerably less than the region disturbed by the Surveyor footpad. The choice of a largest meteoroid diameter will determine, on the average, a maximum time span during which disturbances took place. Let us for simplicity assume homogeneous mixing of rock ejecta to depth L. Let us assume erosion of the rocks of area A_1 at rate $\dot{z}_1$ and homogeneous deposition of rock fragments all of diameter D at rate $\dot{z}_2$ on area A_2. If the rock fragments are deposited locally and do not participate in a net transport of mass away from the region, then $\dot{z}_2 = \dot{z}_1 A_1 / A_2$ and the local soil surface is <u>rising</u>. On the average, a rock fragment then spends a fraction D/L of the time exposed at the surface and is buried beyond depth L after an elapsed time $L/\dot{z}_2$. The net exposure time at the surface for the particle, $D/\dot{z}_2$, is independent of the assumed mixing depth, L.

If we now introduce an assumption of darkening of particles exposed at or nearly at the surface, we see that darkening should be substantially complete in a net time increment of $D/\dot{z}_2$ if a dark soil is being formed from darkening rock fragments in an upper transition layer of thickness L.

If we set the darkening time $\tau = D/\dot{z}_2$, we discover that two of the factors, D and τ , are presumably governed by exogenic or at least moon-wide factors while $\dot{z}_2$ is related to the magnitude of the local rock fragment source. This fact suggests that the low albedo near Surveyor 1 is related to the weakness of the source of rock fragments in that vicinity and that higher albedos in other lunar features might be due to more rapid deposition of rock fragments or to a net transport of surface material that exposes undarkened material.

Let us estimate the darkening time τ from the Surveyor 1 data. For the erosion rate on the rocks, $\dot{z}_1$, let us adopt the erosion rate inferred for chondritic meteorites colliding with interplanetary dust as given by Fisher,[5] 10^{-6} cm/yr. Actually, two partially compensating corrections should be made to this erosion rate. Chondrites spend time in the asteroidal region where dust densities are probably higher. On the other hand, they are not subject to erosion by secondary and tertiary impacts as is the moon. But those secondary impacts on lunar rocks come mostly from primary impacts occurring in the finely divided material between the rocks, so a realistic correction for secondary effects would require better knowledge of the soil properties and of cratering in lunar soil, in particular. The dust density in the

asteroid region is highly uncertain so that correction of the chondritic erosion rate to obtain the erosion by primary impacts at 1 A.U. would be similarly speculative. The dilution factor for the rock fragments, A_1/A_2, is about 2 to 5% near Surveyor 1, depending on how many of the angular blocks at the surface are considered to be primary sources of soil material (which in turn depends upon how ancient is the soil exposed by the footpad). Therefore $\dot{z}_2$ = 2 to 5 x 10^{-8} cm/yr. If we take D = 10^{-2} cm from our polarization studies,[6] we obtain $\tau = D/\dot{z}_2 \approx$ (2 to 5) x 10^5 yr. This time for substantial darkening is probably an overestimate. We therefore emphasize that the darkening time is already very short geologically.

In the above discussion, brightening of the exposed surface could be substituted for darkening provided 1) the rocks of the moon are intrinsically very dark, 2) there is a mechanism which makes exposed surfaces of dark rocks become light, and 3) meteorite erosion is removing the brightened material from the region viewed by Surveyor 1. In discussion of this possibility one can offer the following remarks: 1) Carbonaceous chondrites are intrinsically very dark and might remain so when finely divided. Dark rocks from terrestrial lavas become too light to agree with lunar albedos after comminution to a fine enough powder to agree with lunar polarization properties. But the origin of the moon is sufficiently uncertain so that its composition could resemble carbonaceous chondrites. 2a) In this vein it might be supposed that energetic hydrogen ions of the solar wind might succeed in selectively removing carbon from a superficial layer of such rocks by forming

a volatile end product that could be removed from the moon. We point out that the layer whose transparency could be increased in this manner could not exceed the thickness to which the ions penetrate, about 1000 Å. Hence a rock would not become appreciably lighter under solar-wind action. 2b) Some other rapid surface bleaching process might be offered. The time scale for darkening or brightening has to be the same, and is very short geologically. Therefore any agent causing brightening would have to be fairly active. A possibility might be shock-induced changes induced by meteoroid impact, though this suggestion encounters the unique difficulty that micrometeorites assume a special role. This follows from the fact that the occurrence of an extensive and dark surface soil has to be accounted for. If we accept that the surface rocks are the remnants of large impacts and that the dark surface soil is derived from the (internally dark) rocks by medium impacts, then it falls to the smallest impacts to produce superficial surface brightening. Somehow large and medium impacts then do not produce brightening but the smallest ones do. 3) It is essential to realize that "brightened" material (rock surface material) is not being accumulated around Surveyor 1 in view of the dark soil. It seems unlikely that meteoritic erosion should be preferentially removing matter from the Surveyor 1 region and accumulating it elsewhere on the moon. Therefore the suggestion of a brightening of exposed surfaces almost certainly implies a net removal of matter from the surface of the moon by meteoritic impact. Such "negative accretion" has been

considered by several writers,[7-9] but poses the general problem of finding special conditions for accretion of the moon and other solar system bodies during formation.

We conclude that the simplest explanation of a dark soil and a darker subsoil amidst light rocks in the Surveyor 1 vicinity is a darkening at the surface of a soil being derived from the intrinsically light rocks. We will show in a later section that carbon accumulation from the solar wind alone could have been adequate to darken roughly a meter of surface soil and additional sources of carbon are suggested. In fact, darkening by carbon at exposed surfaces seems inescapable on the moon so that any hypothesis of brightening of exposed surfaces must be able to overcome the carbon darkening.

Finally, let us consider some implications of the above model for other regions of the moon when additional data are taken into account. The model predicts higher albedos where either more rock is exposed or where surface material is being preferentially lost due to horizontal transport. A third possibility for high albedos is that soil slumping introduces overturning of the stratified soil on a vertical scale large enough to expose substantial amounts of undarkened material.

Ray craters emplaced in mare regions are younger than the mare and often outstandingly bright. Many of these bright features are also anomalous thermally[10] and in radar reflectivity[11] in a way suggesting substantially more surface rocks than in mare regions. Our

erosion model suggests that a surface soil will be less fully developed in such youthful features, as other authors have also suggested,[10-12] but it should be rapidly forming. Copernicus, which is included in this class of ray craters, was shown by us[13] to have a photometric function indistinguishable from other regions, including mare surfaces. Therefore the optical properties of Copernicus are already dominated by a surface powder. We know of no lunar feature that has been shown to be photometrically anomalous so a surface powder must develop rapidly in all lunar craters.

The high albedos of extended regions like the highlands pose a more difficult problem. Highland regions have relatively few thermal anomalies during an eclipse and are considerably cooler than maria.[10] This points to a well-developed surface soil. But uplands have a higher radar reflectivity than the maria,[11] which could be due to rocks near the surface or roughness, or both. Ranger 9 and Lunar Orbiter photographs have revealed that most positive relief features (crater walls, central mountains, etc.) are peculiarly free from craters in the 100 to 1000 m diameter range. Orbiter photographs show some evidences for strong slumping on steep slopes. A more common slumping on a smaller scale could be the cause for the paucity of 100 to 1000 m craters. Downhill transport of erosion breccia would be difficult to distinguish from extensive small-scale slumping without detailed study. Whether due to slumping or denuding of slopes by erosion, the Lunar Orbiter photographs reveal most graphically that strong slopes of kilometer dimensions

usually have high albedos. On the other hand, it may be germane

that Lunar Orbiter 2 photographs reveal the extreme rarity of posi-

tive relief objects (rocks?) having dimensions of 1 to 10 m on either

maria or nearby mountain slopes. Further study of these sources of

information is needed before a clear picture can emerge for the older,

more complicated regions. Nevertheless, the higher albedos of strongly

sloping regions are consistent with a model of erosion-produced sur-

face powder being darkened by some moon-wide process.

III. THE CONTROVERSY OVER DARKENING BY CONTAMINATION

The substance of the two papers by Nash and by Greer and Hapke
was that darkening of powders in solar-wind simulation experiments might
be dominantly due to carbon contamination arising from imperfect vacuum
conditions. Greer and Hapke reported concentrations in excess of 10^4
ppm carbon and 10^2 ppm iron in irradiated high purity powders of quartz
and alumina. Analysis was based on detection of characteristic x-rays
excited by an electron beam microprobe. The iron arose from sputtering
in the ion source, but the carbon must have come from carbon-carrying
volatile in the vacuum system. Darkening by a mechanism in addition
to contamination buildup might be indicated by the circumstance that
their helium-irradiated natural quartz was strongly darkened (carbon
$< 10^3$ ppm) while hydrogen-irradiated alumina (carbon $> 10^4$ ppm) was the
least darkened of six samples studied (3 materials bombarded by hydrogen
or helium). However, the existence of another mechanism is uncertain
without further study because carbon detection might be sensitive to
the selection of powder particles (the number of particles studied was
not stated nor was assurance given that the particles were definitely
only from the topmost sample layer) and to operating procedures with the
microprobe (where carbon films also are known to build up). Even if no
objection is sustained, the carbon might be in differing electronic
states in the differing samples such that optical absorption per atom
is quite different.

The darkening of rock powders was found by Nash to be slight in
an ion-pumped system employing a low intensity beam of mostly H^+ ions
(few H_2^+, H_3^+). In contrast, the results published by Hapke[14] and
us[15] indicated marked darkening of similar rock powders in oil-
pumped systems employing high-intensity fluxes of mostly polyatomic
hydrogen ions and similar ion doses. The evidences for carbon con-
tamination given by Nash included: 1) C-H absorption in the infrared
for material scraped from an irradiated oligoclase crystal, 2) high
carbon content detected by α-particle backscattering from a solid
dunite sample that had been irradiated by mass-separated 10 keV pro-
tons. Other circumstantial evidences were given which were not con-
clusive. Darkening occurred adjacent to an O-ring in the vacuum cham-
ber; the cause of the darkening was not stated; the beam was not di-
rected to that area. Darkening on the oligoclase crystal disappeared
after an unstated period at 600°C in O_2 as though carbon were oxidized
to CO and CO_2. But if the film were aluminum from his ion source canal,
then the film would also disappear (become transparent) upon oxidation.
In the system of Nash, elastomer O-rings are sources of carbon-carrying
gas molecules. An oil-sealed mechanical forepump used in roughing is
a less likely source as it is used with a liquid nitrogen cold trap.

In the three investigations of ion-induced darkening of powders
which are to be published, darkening depended on composition in an
analogous way. Pure quartz or alumina darkened relatively little in
comparison to powders of naturally occurring mineral or rock. Nash
mentioned in his paper that the dependence of the rate of darkening

upon composition may be explained in terms of contact catalysis of the decomposition of hydrocarbons on the powder grains, citing Griffith and Marsh.[16] They state that oxide surfaces are much less active as catalysts than metal surfaces and consequently are normally used at higher temperatures. The reason is that only a few percent of the surface is active in catalysis. The (acidic) active sites are less susceptible to poisoning than are metal surfaces, but they can be obscured by the product carbon (a carefully studied problem of the oil industry). They state in relation to hydrocarbon cracking on non-acidic catalysts, "Transitional metals such as iron and nickel cause extensive breakdown to coke and gaseous products through dissociative adsorption and hydrogen transfer reactions." Iron is also used as a catalyst when hydrogen and carbon monoxide are transformed to a wide range of hydrocarbons and alcohols in the Fischer-Tropsch process. Nash then suggested that the higher iron and magnesium oxide contents of rocks, especially of basic rocks, caused the more rapid darkening of rock powders. We support his suggestion tentatively.

The suggestion of catalysis is incomplete without a discussion of how much carbon deposition is expected, where the deposit is located, and the expected optical effects. In the presence of sputtering, carbon cannot accumulate in great amounts on the bombarded surface. For our paper we estimated stopped carbon ions in the solar wind with solar abundance would accumulate a saturation dose of 1.5×10^{16} carbon atom/cm^2 in a SiO_2 layer about 10^{-5} cm thick (less than 2 at. %). We have found

that glass containing 1 at. % of carbon is indeed very absorbent. But the very small thickness of the layer in which this carbon is accumulated at the energies of solar-wind ions (~10 keV for C^{6+}) is highly transparent. This is in accord with our observation that solid quartz or glass is negligibly darkened in sputtering with Hg^+ and H_2^+ ions of up to 2 keV in conditions where carbon ions were certainly a strong impurity. But if the carbon is sputtered or otherwise transferred to a portion of an adjacent powder grain that is not being bombarded, it can be incorporated with silicaceous material into a thick and highly stable layer. The concentration of carbon in a built-up layer might be at least approximately equal to that in the 10^{-5} cm layer in which ions are stopping if transfer from the bombarded surface to the unbombarded one is principally by sputtering. Direct experiments on whether some carbon is lost to volatile substances through surface reactions should be made.

But in fact, a gain in carbon beyond that due to the solar wind should occur. Trapping of carbon-carrying volatiles from the tenuous lunar atmosphere is to be expected in a building-up layer. The source of carbon-carrying gases could be meteoroid impact explosions or volcanic events. They could be trapped by either of two routes. First, the sputtering of the lunar surface should prepare many surface sites (atomic in size) where carbon-carrying neutral molecules will be catalytically transformed to carbon or extremely non-volatile substances. Some such sites will lead to immediate entrapment of carbon by the sputtered flux of lunar surface atoms. Other sites are exposed and will be

disturbed by ion bombardment (within a time period measured in years).
Some of this carbon will then be moved to stable sites. Second, some
carbon-carrying molecules emitted into the lunar atmosphere will be
dissociated by solar light into radicals and ions that will strongly
interact with the lunar surface upon impact.

Many uncertainties exist in a quantitative estimate of lunar op-
tical effects due to carbon. We want to remark that carbon in the
solar wind alone could have contributed quite markedly to lunar dark-
ening. At 10^5 C^{6+}/cm^2-sec for 4×10^9 yr, one accumulates 10^{22} carbon/
cm^2. Since a few atomic percent carbon in a 5μ layer on 100μ parti-
cles constitutes an approximate model for lunar surface darkening in
terms of presently available data, one predicts marked darkening of
about 10^{25} silicate atoms/cm^2 or about 400 gm/cm^2. This estimate
should be decreased if carbon in the solar wind is below the solar
abundance (6×10^{-4} from flare and spectroscopic data[17]), or if
carbon ions are deviated around the moon some of the time due to col-
lective effects in the magnetized solar wind. We discussed both of
these decreases previously[18] and concluded that they might together
decrease the estimate by one or two orders of magnitude. Our point
is that darkening due to carbon in a lunar surface layer probably can-
not be ignored even if additional darkening mechanisms exist, such as
the metal enrichment that we have expected throughout our investigating
period.

In favor of the role of catalytic cracking of carbon molecules in
past simulation experiments, we remark on the slight darkening of pure

SiO_2 and Al_2O_3 compared to the marked darkening of a simple 1:1 mixture of the two powders observed by Hapke[14] and recently by us in a dish having powders in 4 quadrants. Ion bombardment is the key in these experiments to maintenance of active sites for catalysis and to contact between the gas and solid phase components. Because of transfer of atoms between powder grains, a mixture of particle compositions results in a composition mixture at the active surfaces. Griffith and Marsh[16] cite a large number of data supporting promotion of catalysis "by a second substance B added to a one-component substance A". In fact, an SiO_2-Al_2O_3 mixture is an especially well-studied example of enhanced catalysis in cracking of carbon molecules by oxide surfaces.

Finally, we indicate existing data that imply an adequate source of carbon contaminant in the experiment of Hapke. In a discussion of contamination of fresh silica by silicone oils, Holland[19] reported the loss of surface wetting by water and the loss of affinity for zinc sulfide after an exposure of 3 to 5 min to silicone 704 at room temperature. This he took to be the time for a monolayer of oil to form on the freshly evaporated silica. This time agreed with the rate of transport of oil molecules to the surface for a published vapor pressure of 5×10^{-9} Torr for that oil at the same temperature, provided that 100% of the oil molecules were adsorbed. If the silica surface was held at 230°C, then the length of time for full coverage to develop increased to 75 min, implying that only about 1/20 of the molecular impacts on the heated silica resulted in chemisorption. Silicone 704 is a tetramethyl tetraphenyl trisiloxane having a molecular weight of 484. Silicone 705 differs from

704 by having one more phenyl group that replaces a methyl group and, in consequence of the greater molecular weight, it has a vapor pressure lower by about a factor of 3. Notice that each adsorbed silicone 705 molecule involves 33 carbon atoms.

Hapke believed the partial pressure of silicone 705 vapor near his sample was considerably less than 1×10^{-10} Torr on the basis of data from a commercial gas analyzer similar to ours. But the parent ion at m/e =546 is outside the mass range of the analyzer so one must know the ionization pattern of the silicone molecule accurately for the particular gas analyzer in order to obtain a pressure determination. In the presence of active pumping, the partial pressure of the silicone oil will be somewhat lower than the vapor pressure. We consider a pressure of 1×10^{-9} Torr as an example. The number of oil molecule collisions in the course of Hapke's bombardment time of 0.95×10^{5} sec was approximately 5×10^{16} cm^{-2}. If we estimate the sticking probability for each silicone molecule on his beam-heated powder sample was 1/33 and remember that 33 carbon atoms are in each molecule, then 5×10^{16} carbon atoms/cm^{2} were adsorbed on the sample surface. The fate of such carbon atoms is, of course, not known in detail. But if most of this carbon were finally incorporated into a silicaceous layer, then such a layer would be 1.5μ thick at 1% carbon content. Since the penetration of the probing electrons is comparable to this thickness, we conclude that even such a minute pressure of contaminating gas can account for the carbon detected. A similar conclusion might follow for CO as a contaminant, since its partial pressure was 3×10^{-9}

Torr. But in this case only one carbon atom is supplied per molecule. Therefore the sticking probability would have to be at least 0.1 to supply enough carbon. The number of active sites for catalysis on pure oxide substances is generally lower than 0.1. On the other hand, CO^+ ions in proportion to the partial pressures of H_2 and CO (ratio of about 3×10^{-5}) would supply an additional 2×10^{16} carbon atoms/cm^2 and oil ions another 20×10^{16}. The sticking probability for atoms supplied in ionic form should be near unity. Therefore impurities in the ion beam, which was not mass-analyzed, should also be considered.

The above estimates are order of magnitude only. But they serve to emphasize that important carbon contamination is extremely hard to avoid in simulation experiments. It is hard to avoid as a factor in lunar darkening as well.

IV. APPROXIMATE LOMMEL-SEELIGER SCATTERING

Hapke[20] offered the suggestion that the brightness of individual lunar features is governed by a relation of the form

$$I(i,\epsilon,\alpha) = L(i,\epsilon)F(\alpha), \tag{1}$$

where the function $L(i,\epsilon)$ is the Lommel-Seeliger function

$$L(i,\epsilon) = (1 + \cos \epsilon \sec i)^{-1} \tag{2}$$
$$= \cos i/(\cos i + \cos \epsilon),$$

and where the second function $F(\alpha)$ was separated into another product

$$F(\alpha) = S(\alpha) \times B(\alpha). \tag{3}$$

The function $S(\alpha)$ describes how an average particle scatters light and $B(\alpha)$ results from shadowing within a tenuous surface of small, opaque particles. Since Hapke gave explicit expressions for $S(\alpha)$ and $B(\alpha)$, he compared the product function $L(i,\epsilon)F(\alpha)$ to photometric data on lunar features but did not test the validity of his factorization explicitly. The recent photoelectric data of Gehrels et al.[21] were re-analyzed[13] to test this factorization with the result that the factorization is, indeed, a valuable approximation for central lunar features. Briefly, $F(\alpha)$ should be symmetric about $\alpha = 0$, so multiplying the intensity data by $L^{-1}(i,\epsilon)$ should result in identical curves for positive and negative phases. A representative result is given in Fig. 1. It may be seen that after multiplication by $L^{-1}(i,\epsilon)$, the
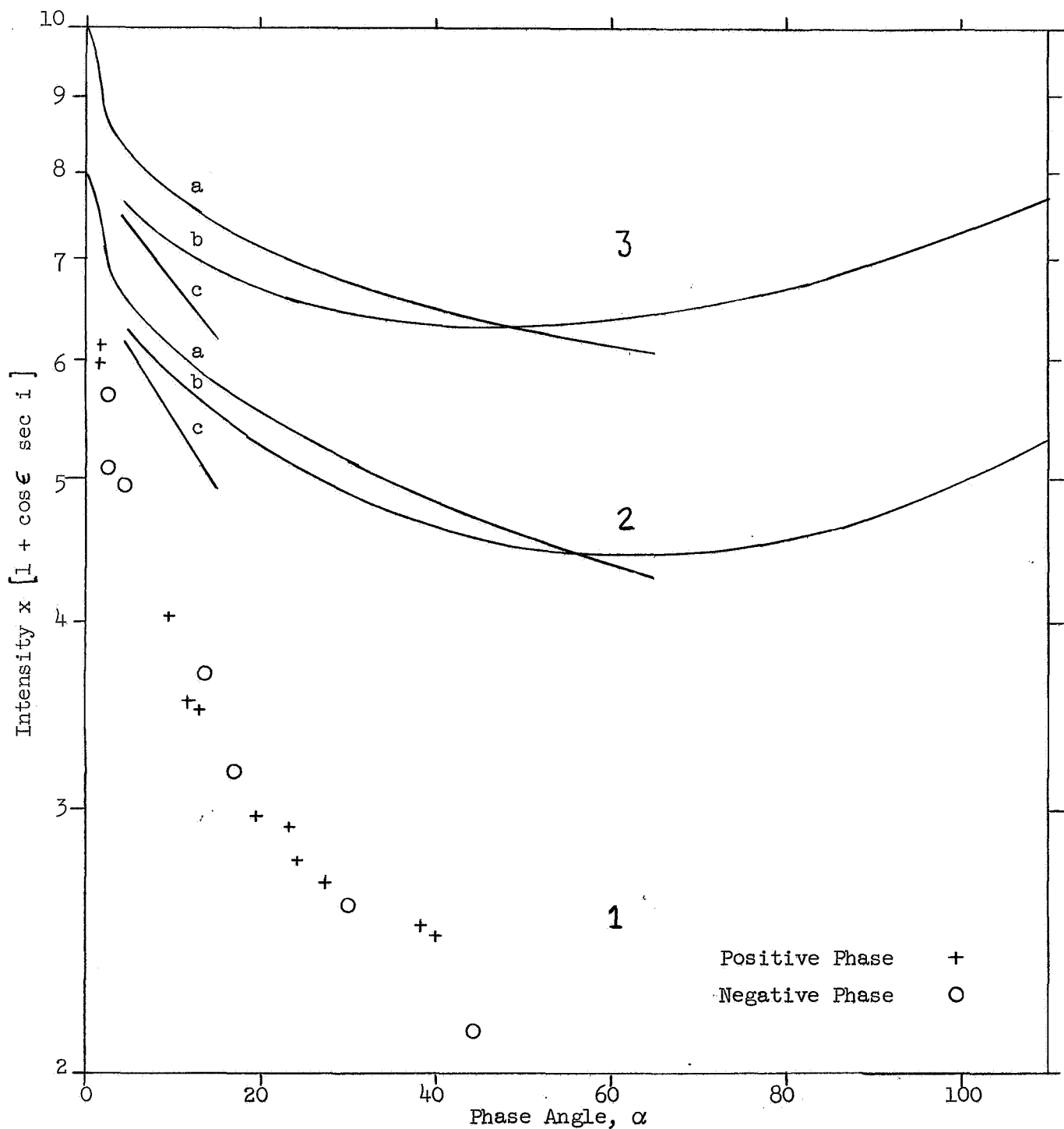
Fig. 1 Photometric functions F(α) in green light of: 1) Mare Tranquillitatis,
latitude 39?3, according to Gehrels, et al. (1964), 2) sifted 10-20 μ
tholeiitic basalt powder, A_n = .107, 3) same powder compacted by shaking,
A_n = .110. Curves (a) are for ϵ = 0°, (b) for ϵ = 50°, (c) for ϵ = 50°
with light source between detector and the surface tangent plane. Curves
(a), (b), and (c) are not arbitrarily placed but are related by limb-
darkening effects. The circled points in (1) correspond to curves (b);
the others correspond to curves (c).

data points on the brightness of a small region in Mare Tranquilli-
tatis for positive and negative phases lie on one curve to the 5% pre-
cision inherent in the data. Hapke[22] later pointed out a diffi-
culty that arises if the moon is considered to be a smooth sphere scat-
tering according to the assumed law. At phases away from full moon the
illuminated limb is predicted to be especially bright. Though no com-
pelling experimental evidence indicates a failure of the suggested
photometric function, Hapke felt that a smooth moon was physically un-
realistic and offered a modification of the function $L(i, \epsilon)$ that affects
the brightness of limb features. Central lunar features are affected
negligibly, particularly near full moon. After considering what impli-
cations for surface compaction follow from Lommel-Seeliger scattering
properties, we will cite some evidence that strongly suggests that
Lommel-Seeliger scattering does in fact fail at the limb.

If a powder surface is a Lommel-Seeliger scatterer in the sense of
(1), then viewing the sample normal to its surface or from an oblique
angle with $\alpha = 0$ should reveal an identically bright sample because
$L(i = \epsilon)$ = constant. In Fig. 1 a sample of basalt powder is shown to
be slightly darker when viewed obliquely. The sample that was sifted
for minimal compaction was 5% darker at $\epsilon = 50°$ than at $\epsilon = 0°$, whereas
the sample that was purposely jarred for compaction was 10% darker at
$\epsilon = 50°$ than at $\epsilon = 0°$. Second, if these surfaces were Lommel-Seeliger
scatterers, the curves in Fig. 1 for $\epsilon = 0°$ and 50° should have the same
shape, and the shape should be independent of the sign of the phase angle.

In this test the sifted surface is again superior to the compacted one as a Lommel-Seeliger scatterer, but the function $I \times L^{-1}$ for $\epsilon = 50°$ becomes progressively greater than that for $\epsilon = 0°$ at large phase angles. Unlike the Mare Tranquillitatis region, which is also viewed obliquely, a reversal of the sign of the phase angle at $\epsilon = 50°$ does not give identical curves for the basalt sample after multiplication by L^{-1}. Therefore the dependence of the brightness of the basalt powder sample in Fig. 1 upon the angles of incidence and emergence is imperfectly described by $L(i,\epsilon)$, though the discrepancies are less than 10%. Darkening of such samples by sputtering invariably improves the agreement between the $\epsilon = 0°$ and $\epsilon = 50°$ curves. The laboratory data of Fig. 1 were measured to 0.1% precision and are free from the error of light scattered from the sample container.

Tests of the accuracy of (1) other than the symmetry of $I \times L^{-1}$ mentioned above are not feasible for observations made from the earth unless $F(\alpha)$ is considered to be known. For example, the darkening of the powder sample when viewed at an oblique angle with a small phase angle would correspond in the lunar case to a slight limb darkening. Qualitatively we know lunar limb darkening is small, but any quantitative statements make some assumption of equality of albedos for similar features. Second, the similarity of the photometric curves $I \times L^{-1}$ for the powder sample at different viewing angles for a range of phase angles near $\alpha = 0$ would correspond in the lunar case to comparing photometric curves for differing ϵ on the same feature. Small

changes in ϵ for a feature do occur because of libration but are experimentally difficult, less than $\pm 8°$. Again, different features can be compared. The data of Gehrels et al. reveal that for 13 lunar features of various types, the variations in the shapes of $F(\alpha) = I \times L^{-1}$ are so small as to be scarcely distinguishable in the interval $5° < |\alpha| < 50°$. Furthermore, if one assumes (1) holds for every lunar area, the brightness of the whole moon is given by

$$T(\alpha) = \left[1 - \sin \frac{\alpha}{2} \tan \frac{\alpha}{2} \log \cot \frac{|\alpha|}{4} \right] \times \bar{F}(\alpha) \cdot \qquad (4)$$

When the Lommel-Seeliger dependence of (4) is removed from the data of Rougier,[23] the mean phase angle function $\bar{F}(\alpha)$ is in very good agreement with all the lunar features of Gehrels et al. with the possible exception of the bright ray crater Tycho. For Tycho the function $F(\alpha)$ seems to decrease less rapidly with increasing $|\alpha|$. This comparison of photometric curves for differing ϵ (on different features) would suggest that all the features are good Lommel-Seeliger scatterers. Indeed, all the features pass the phase-angle symmetry test of Fig. 1 equally well.

The porosity of the sample surface studied in Fig. 1 was 80 to 90% after sifting and about 50% after compaction. The higher porosity of the sifted surface did improve behavior as a Lommel-Seeliger scatterer. But the improvement in the phase-angle symmetry test was not marked. Certainly the sifted surface was quite inferior in this test to the region of Mare Tranquillitatis studied by Gehrels et al. We

admit that such evidence might require an especially porous lunar surface but caution against such an interpretation yet. We have found that a powder having dull facets (greenstone, see below) is considerably superior to our basalt powder sample as a Lommel-Seeliger scatterer, even in a size range which is self-compacting, 20-44μ. But as we shall presently show, the compacted greenstone powder also exhibits a backscatter peak which is too strong to agree with the moon whereas basalt is too weak. Unfortunately, we have not yet studied a powder having a backscatter peak in close agreement with the moon so that we do not know if such a powder must be in a very porous state in order to exhibit the Lommel-Seeliger scattering found for central lunar features.

Very few of the data of Gehrels et al. extend beyond $|\alpha|$ = 50° but the data of Rougier for the whole moon extend from α = -130° to α = +150°. We want to point out a feature of Rougier's data that requires a modification of Lommel-Seeliger scattering of the sort suggested by Hapke.[22] If one assumes the validity of (1) and a smooth lunar surface, (4) follows. But when the term in (4) arising from assumed Lommel-Seeliger scatteringis removed from the data of Rougier,[23] the function $\bar{F}(\alpha)$ continues to decrease exponentially with phase angle in the whole range studied by Rougier. For example, $\bar{F}(120°)/\bar{F}(5°)$ = 0.155. In contrast to such a great decrease, our analogously obtained function $I \times L^{-1}$ in Fig. 1 and in other comparable data decreases only slightly beyond $|\alpha|$ = 60° or in fact increases beyond 60°. For powders having albedos

in the lunar range, $F(120°)/F(5°)$ is 0.5 to 1. For much darker powders, smaller ratios are found. But the decline in $F(\alpha)$ up to $|\alpha| = 60°$ is then too rapid and is followed by a slow decrease to $|\alpha| = 120°$.

Since the illuminated limb of the moon is not smooth but appears to be a series of tilted surfaces to an observer, the assumption of (1) in unmodified form overestimates the coefficient of $\bar{F}(\alpha)$ in (4). Therefore $\bar{F}(\alpha)$ is underestimated when (4) is inverted. According to Hapke, the relative error will increase with phase, as is noted here. We conclude that the strongly declining brightness of the whole moon beyond $|\alpha| = 60°$ requires important shadowing effects in a surface macrostructure. Careful measurements of the brightness variations of limb features at large phase angles should permit separation of the effect of shadows due to mountainous terrain from shadows in the macrostructures resolved in the Luna 9 and Surveyor 1 photographs. One would assume an $F(\alpha)$ based on laboratory data, provided such data were reproducible under suitable experimental conditions. We have not yet carried out such a program. In any case, it should be clear that no evidence on the lunar surface compaction is forthcoming from photometric data at large phase angles because the macrostructure is not intimately known.

The main change that this section represents is that effects of a rough macrostructure must be incorporated into that part of the lunar photometric function that depends upon the angles of incidence and emergence, namely into $L(i,\epsilon)$ in (1). We had previously supposed that this Lommel-Seeliger factor was so well-demonstrated for the lunar case that

residuals between the lunar function and laboratory samples had to be found in $F(\alpha)$. But the features studied by Gehrels et al. were distant from the limb regions where the Lommel-Seeliger factor must be modified.

V. COMPLICATIONS IN THE LUNAR PHOTOMETRIC FUNCTION ARISING FROM MULTIPLE SCATTERING

A. Color Changes with Phase at Moderate Phase Angles

At small phase angles, the effects of shadows in a macrostructure should become negligible in comparison with shadows in a very porous microstructure. The theory of Hapke[20] was intended to treat this case of small phase angles, but it contains some mathematical flaws in the final steps which will be discussed elsewhere. Irvine[24] gave a more exact analysis of shadowing effects, including multiple scattering effects, drawing from theories stimulated by observations of the rings of Saturn. But the theory is not complete without assuming a scattering function $S(\alpha)$ for an average particle and performing numerical integrations of several integral equations. Besides, the approximation of geometric optics was made so that a softening of shadows by diffraction could be treated only qualitatively. The brightness variations of a lunar feature could depend upon photon frequency because of multiple scattering and diffraction. We now cite lunar and laboratory photometric observations that suggest the first of these corrections to primary scattering has been detected in two ways and should be incorporated into an adequate theory. We will conclude that the lunar photometric function is compatible with scattering from a powder surface but that a low-density surface is not necessarily required.

Gehrels et al. observed that the lunar backscatter peak is stronger at higher photon frequencies (lower particle albedos). What they noted was that the moon becomes more reddish with phase according to the relation (corrected for the color of sunlight)

$$(U-G) = +0.496^m + 0.0036 \ |\alpha| \ .$$

This indicates that the moon is brighter in green light than in ultra-
violet light by about 0.5 stellar magnitudes (by 1.62) at full moon and
that the ratio is increased by another 14% at $|\alpha|$ = 40°. Wildey and
Pohn[25] did not notice such a tendency in the shorter frequency inter-
val from blue to green, probably because of excessive scatter intro-
duced by inadequate corrections for atmospheric extinctions. They noted
a correlation between the mean color of 25 lunar features observed on
14 nights in 1962-1963 and the mean extinction coefficient on the res-
pective nights. We therefore calculated the mean value of (U-V) for
each of their runs on 25 features (1 or 2 runs per night) and obtained
a correlation relation to the extinction coefficient X in U-B of the
form

$$\overline{U\text{-}V} = 1.517 - 0.765X \ .$$

After correction of each $\overline{U\text{-}V}$ to constant extinction coefficient, we
found that these data do support a reddening with phase amounting to
0.0036 mag/deg in the U,G interval of Gehrels et al.). The dependence
of color on phase might be uncertain by a factor of two according to
these badly scattered data, but the effect seems real. Concerning our
treatment of the data of Wildey and Pohn we remark that it seems de-
sirable to study the largest frequency interval, U-V, to take advantage
of the greater albedo contrast, that the extinction coefficient for U-B
was the only one published, and that the mean color of a group of fea-
tures should exhibit a phase dependence common to each of them. Coyne[26]
studied the differences in color of a large number of lunar features at
various phases and found no phase variation in the lunar color differences.

Apparently the phase dependence of lunar color exhibits the same kind of unanimity that the lunar color does. We indicate below why this should be so.

In Fig. 2 we show that a reddening with phase of the proper magnitude is present in scattering from a powder, in this case from a metamorphic rock commonly called greenstone. This powder has relatively opaque particles with rough facets, but is otherwise undistinguished. The steeper descent of $F(\alpha)$ for the unsputtered or sputtered samples when measured in ultraviolet light instead of green light amounts to an increase in the ratio of brightnesses of roughly 14% at $|\alpha| = 40°$. The fact that the symmetry of the curves in Fig. 2 is not identical in the different colors on the same surface resulted from moving the sample between changing filters in order to make the albedo determinations. Therefore slightly different sample areas were viewed. The sharper decrease in $F(\alpha)$ as photon frequency increases has been confirmed for all reddish surfaces so far studied, but the phase dependence of the color vanishes for gray powders like graphite, SiC, or MgO. By studying a fixed area of the sample, the phase coefficients of the color of these powders in several size ranges were found to be less than 10^{-4} mag/deg in our blue-to-red interval.

The way to see that this dependence of the photometric function upon color is due to multiple scattering and is negligibly due to diffraction is to notice that the extent of multiple scattering depends upon albedo but not upon photon frequency. Therefore at different
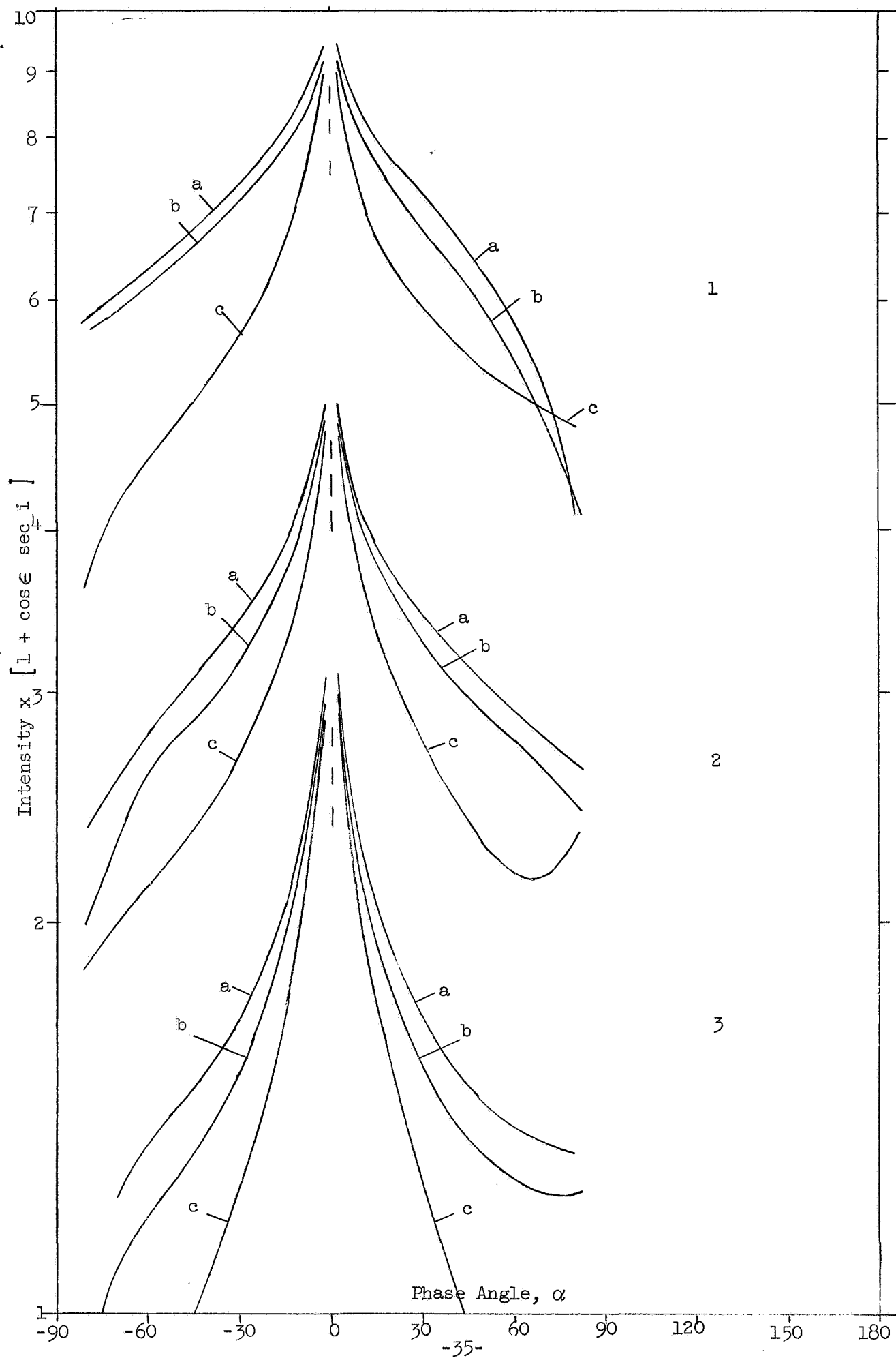
Fig. 2 Photometric function F(α) at $\epsilon = 0°$ for sifted 0-20μ greenstone powder. The function depends markedly on the wavelength of the scattered light. a) Red light, b) Green light, c) Ultraviolet light. 1) Unsputtered, 2) 10^4 equiv. yr., 3) 10^5 equiv. yr.

frequencies for which the albedo is the same, the measured photo-metric function shapes should be the same if multiple scattering is the cause of this color-phase effect. The photometric function is independent of photon frequency for the gray powders mentioned above. The effect can also be seen in Fig. 2. The unsputtered sample has an albedo of 0.18 in ultraviolet light (curve 1c); after sputtering for the equivalent of 10^4 years, the sample has an albedo of 0.13 in red light (curva 2a); the function shapes are closely similar despite the difference in photon frequency. The analogous curves 2c and 3a in Fig. 2 are also closely similar to each other; the respective albedos are 0.068 and 0.062. We find a similar result for larger particle sizes. Since the surfaces are compacted for the larger particle sizes, this effect therefore does not require an underdense surface structure. In particular, one should not hope that the dependence of lunar color upon phase at $|\alpha| > 3°$ will imply anything about the lunar surface compaction.

The main change in this section was the inclusion of data from Wildey and Pohn, a reference pointed out to us by a referee. In Figs. 3 and 4 we give the analysis that led us to the conclusion that these data do indeed support a dependence of lunar color upon phase. We also examined whether there was any dependence of the $\overline{U-V}$ to X correlation on the angle of elevationof the moon. This was motivated by the idea that possibly the extinction corrections had simply been carried out incorrectly in the computer program. But no obvious dependence on the angle of elevation of the moon appears.
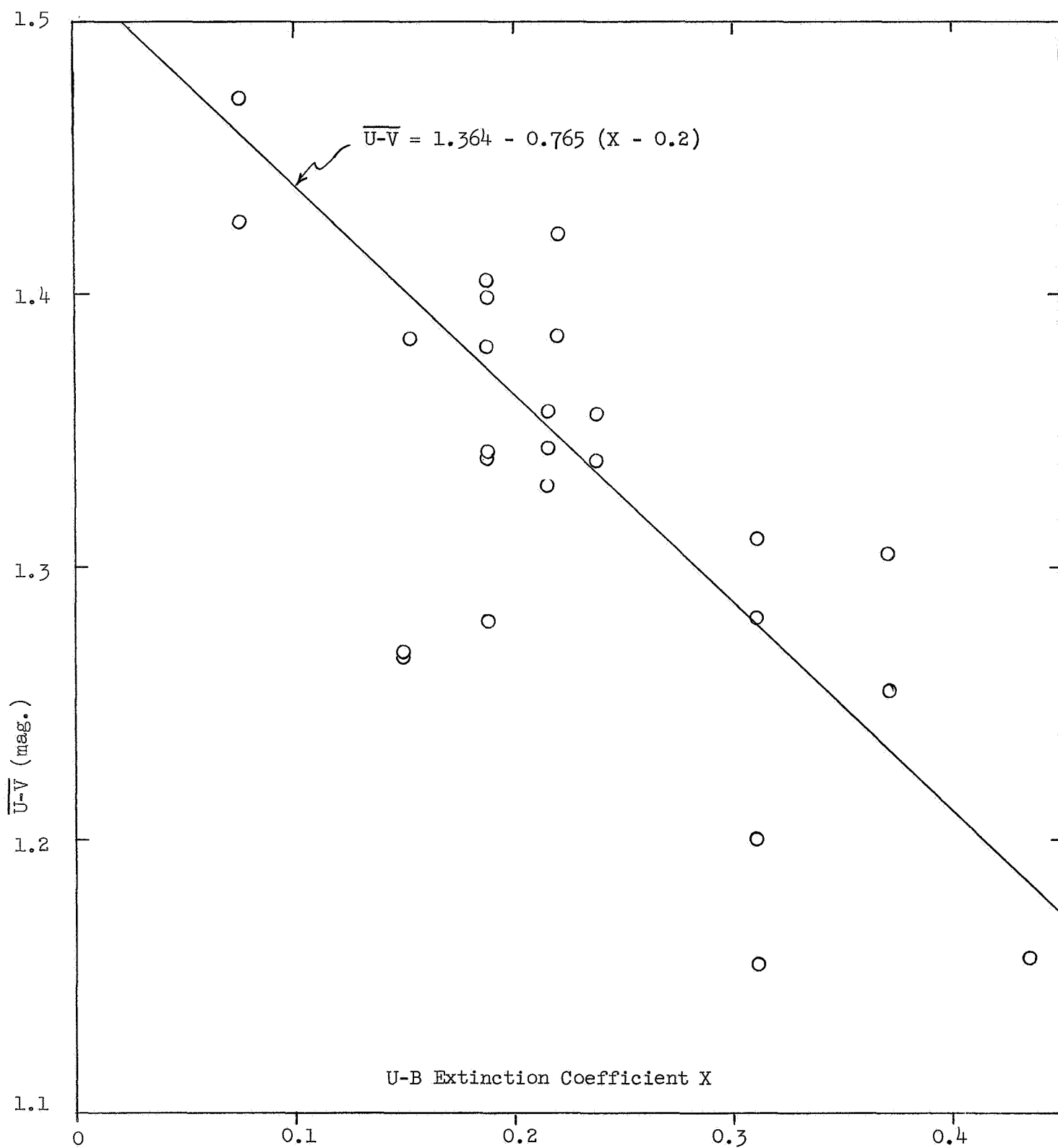
Fig. 3 Correlation between U-B extinction coefficient for an observing night and the mean U-V brightness ratio for 25 lunar features as given by Wildey and Pohn.[25]
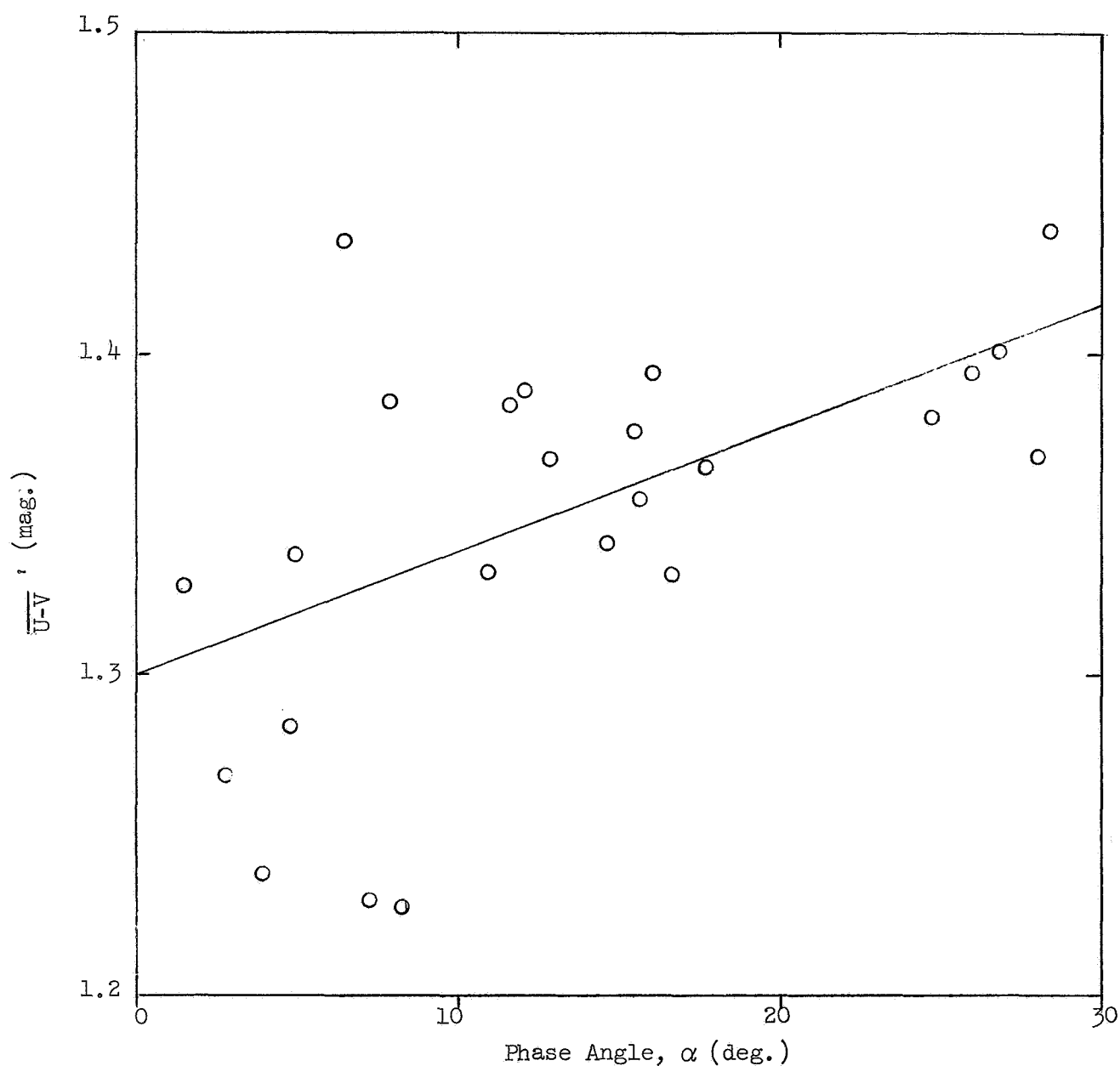
Fig. 4 Variation of apparent lunar color with phase angle for 25 lunar features studied by Wildey and Pohn.[25] The ratio of brightnesses in green and ultraviolet was first averaged over the features ($\overline{U-V}$) and then corrected to constant U-B extinction according to the linear regression of Fig. 3. The corrected mean color U-V' is plotted at the mean phase angle occurring during the observation of the 25 features. The plotted curve has the slope suggested by Gehrels et al.[21] for this photon frequency interval, 0.0036 mag./deg.

Another change was our acceptance of the analysis of Irvine[20]
following private correspondence with Hapke, who succeeded in clari-
fying the physics involved.

B.   The "Opposition Effect"

In Fig. 1, the photometric function for the basalt powder sample
is seen to have two fairly distinct regions: an abrupt drop in bright-
ness to $\alpha = 3°$ followed by a more gentle decline at larger phase angles.
Most of the breadth of the peak at small phase angle is due to the
angular resolution of the reflectometer: $\Delta \alpha = 1.0°$, divided into
$\Delta \epsilon = 0.3°$ and $\Delta i = 0.7°$ in approximately rectangular responses. The
laboratory data in Fig. 1 for $|\alpha| \geq 1°$ was measured in the usual way
with the reflectometer. The data for $|\alpha| \leq 5°$ were checked with a
beam-splitting arrangement and the point for $\alpha = 0$ was thus added.

The steep decrease of the lunar $F(\alpha)$ for $0 < |\alpha| < 5°$ as $|\alpha|$
increases was first established by Gehrels et al., then extended to
additional features by van Diggelen,[27] who also reviewed the earlier
data in which this oppostion effect was suggestive. The calibration
of van Diggelen depended upon that of Gehrels et al., but in re-
analyzing their data we found such a large scatter for $|\alpha| < 3°$
that we can only estimate $1.25 < F(0°)/F(5°) < 1.5$. Variations in
the opposition effect with type of lunar feature cannot be confidently
established from these data. By comparision, the ratio $F(0°)/F(5°)$
was estimated to be in the range 1.6 to 2.2 by Gehrels et al. and
van Diggelen, which seems to be an overestimate. The ratio is 1.23

for the sifted powder of Fig. 1 and only 1% less for the compacted

sample. Both of our ratios are apt to be underestimated because our

angular resolution $\Delta\alpha = 1.0°$ is about three times poorer than that

of the sun-moon-observer system.

According to Franklin and Cook,[28] Saturn's rings exhibit a

marked opposition effect. The ratio $F(0°)/F(5°)$ was 1.49 in visual

frequencies and 5% greater in blue light. The change in brightness

with phase is particularly rapid for $|\alpha| < 2°$ and the difference in

$F(0°)/F(5°)$ for blue and green light develops in this angular range.

For $|\alpha| > 2°$ the rate of change in brightness is 0.036 mag/deg and

this slope is the same for the two frequencies, within the experi-

mental uncertainties. It is nearly the same as those for the moon

(0.028 mag/deg) and Mercury (0.038 mag/deg) according to Harris.[29]

For $|\alpha| > 2°$ the color of the scattered light is (B-V) = 0.86 mag,

of which 0.63 mag results from the color of the sun. Since (B-V) =

0.84 mag for the moon according to Gehrels et al. the ring particles

are slightly more reddish than the moon's surface. Since $\alpha = 5.5°$

is the largest accessible phase angle for Saturn as seen from the

earth, a color-phase relationship of 1.7×10^{-3} mag/deg (as given by

Gehrels et al. for the moon in B-V) would not be detectable in Saturn's

rings beyond $|\alpha| = 2°$, even if it existed. Franklin and Cook attached

great importance to the color-phase relationship for $|\alpha| < 2°$ as a feature

of the oppostion effect for Saturn's rings and sought to account for

it with two alternate models involving the presence of micron-size

particles in the rings. The opposition effect itself was assumed to

result from shadowing by independent particles. Because the angular

diameter of the sun is only 3.4' at Saturn, the dust cloud in either

model must be quite dense. The ring thickness, surprisingly, is only

of the order of 10 cm according to this interpretation.

We wish to remark that the opposition effect which has been es-

tablished for the moon, Saturn's rings, and certain asteroids[30-31]

is probably a multiple-scattering phenomenon involving shadows in a

porous substance (powder?). In observations to be reported in more

detail elsewhere, we have found that the color of the light back-

scattered from a powder changes abruptly at small phase angles just

as for the rings of Saturn, provided the powder is reddish. But for

the gray powders mentioned above there is no color-phase relation-

ship for large or small phase angles. We surmise that the fact that

this effect reveals itself at very small phase angles is related to

the high reflectivity of any dielectric surface at glancing inci-

dence and emergence. If so, the occurrence of the opposition effect

only requires a surface in which are located some pores that are rather

deeper than they are wide. Since the opposition effect is especially

marked in forests and grassy fields to an observer in an airplane,

the mere occurrence of an opposition effect does not require micron-

size particles, obviously. Further experiments with better angular

resolution that we have used and more lunar observations are needed

before any decision can be made on whether the lunar opposition

effect requires an especially porous surface. We do expect, however,

that the lunar opposition effect will exhibit a color-phase pheno-
menon comparable to that observed in Saturn's rings.

The foregoing interpretation of the opposition effect is announced
for the first time in the paper to be published. We here give a few
remarks. The reddening with phase in the opposition effect and at
larger phase angles on reddish surfaces indicates the gradually in-
creasing importance of multiple scattering. But the changes in color
appear to be minor beyond about $\alpha = 40°$. The opposition effect on
highly transparent crystals like MgO might be complicated by the pre-
sence of coherent reflection toward the light source.

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